2.95-MeV and 14.8-MeV Neutron-Induced Fission of Th^{232} †

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Fission yields were determined radiochemically for sixteen mass chains from neutron-induced fission of Th^{232} using 14.8-MeV neutrons and for ten mass chains using 2.95-MeV neutrons. Fine structure in the mass yield curves in the region $A=131$–135 was observed at both neutron energies with the peak in the fine structure occurring at $A=134$.

I. INTRODUCTION

FINE structure in the mass yield curves of several heavy nuclides has been observed by many investigators.[1–11] However, no such fine structure has been reported for the fission of Th^{232} with neutrons of discreet energies. The only investigations reported to date of Th^{232} fission induced by 3- or 14-MeV neutrons are those by Vlasov *et al.*[12] and by Turkevich and Nidday.[13] However, in neither of these studies were the fission yields for the consecutive mass chains $A=131$–135 determined, and consequently observation of fine structure in this region was not possible. Thus, the present investigation was undertaken in an effort to observe nuclear shell effects on fission yields near the closed neutron shell, $N=82$, and to further characterize the mass yield curves for Th^{232}.

$Th(NO_3)_4$ was irradiated with 14.8±0.2-MeV neutrons and with 2.95±0.08-MeV neutrons, and fission yields were determined radiochemically for mass numbers 89–92, 99, 111–113, 115, 131–135, 139, and 140.

II. EXPERIMENTAL

Target Material

The Th^{232} used in this work was reagent-grade thorium nitrate ("Baker's analyzed").

Irradiation Techniques

The Th^{232} samples were irradiated with neutrons produced by the University of Arkansas 400-kV Cockcroft-Walton positive ion accelerator.[14] In one series of irradiations, 14.8±0.2-MeV neutrons produced by the $T(d,n)He^4$ reaction were used. In another series, 2.95±0.08-MeV neutrons produced by the $D(d,n)He^3$ reaction were used. Total neutrons yields obtained were 10^{10}–10^{11} neutrons sec^{-1} at 14.8 MeV and 10^8–10^9 neutrons sec^{-1} at 2.95 MeV.

After irradiation, the length of which was varied from about ten minutes up to several hours depending upon the half-lives of the particular nuclides for which fission yields were to be determined, the sample of thorium nitrate was dissolved in warm dilute HNO_3 containing appropriate amounts of inactive carriers of the various elements to be isolated. After cooling, this solution was diluted to a known volume, and aliquot portions were removed for chemical separation at various lengths of time after the irradiation. Mo^{99} was isolated from each target as a reference nuclide after allowing sufficient time for the decay of the short-lived molybdenum isotopes. The chemical separation procedures employed were standard methods.[15–19] To eliminate the contamination of the barium samples by radium isotopes, the thorium targets used for the barium determinations were separated from radium immediately before irradiation, and a blank of the radium activity was counted and subtracted from the barium data.

Counting Techniques

Both gross beta decay counting and gamma spectrometry techniques were used. The beta counting was done in a 2π end-window methane-flow proportional counter. Gamma-ray spectra were taken by means of NaI(Tl) detectors and a 200-channel pulse-height analyzer.

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[1] H. G. Thode and R. L. Graham, Can. J. Research **25A**, 1 (1947).
[2] J. MacNamara, C. B. Collins, and H. G. Thode, Phys. Rev. **78**, 129 (1950).
[3] R. K. Wanless and H. G. Thode, Can. J. Phys. **33**, 541 (1955).
[4] C. W. Stanley and S. Katcoff, J. Chem. Phys. **17**, 653 (1949).
[5] D. R. Wiles, B. W. Smith, R. Horsley, and H. G. Thode, Can. J. Phys. **31**, 419 (1953).
[6] W. H. Fleming, R. H. Tomlinson, and H. G. Thode, Can. J. Phys. **32**, 522 (1954).
[7] W. H. Fleming and H. G. Thode, Phys. Rev. **92**, 378 (1953).
[8] F. T. Ashizawa and P. K. Kuroda, J. Inorg. Nucl. Chem. **5**, 12 (1957).
[9] K. M. Broom, Phys. Rev. **126**, 627 (1962).
[10] E. P. Steinberg and L. E. Glendenin, Phys. Rev. **95**, 431 (1954).
[11] D. M. Wiles, J. A. Petruska, and R. H. Tomlinson, Can. J. Chem. **34**, 227 (1956).
[12] V. A. Vlasov, Y. A. Zysin, I. S. Kirin, A. A. Lbov, L. I. Osyaeva, and L. I. Sel'chenkov, AEC-tr-4665, 1960 (unpublished).
[13] A. Turkevich and J. B. Nidday, Phys. Rev. **84**, 52 (1951).
[14] W. L. Bronner, K. W. Ehlers, W. W. Eukel, H. S. Gordon, R. C. Marker, F. Voelker, and R. W. Fink, Nucleonics **17**, No. 1, 94 (1959).
[15] E. M. Scadden and N. E. Ballou, NAS-NS-3009, 1960 (unpublished).
[16] D. N. Sunderman, NAS-NS-3010, 1960 (unpublished).
[17] J. Kleinberg and G. A. Cowan, NAS-NS-3005, 1960 (unpublished).
[18] D. N. Sunderman and G. W. Townley, NAS-NS-3047, 1961, (unpublished).
[19] E. K. Hyde, NAS-NS-3004, 1960 (unpublished).

All samples were mounted by using the filter-stick technique to provide an evenly distributed precipitate over a well-defined area (3.14 cm^2). Beta counting was performed using a saturation-backscattering thickness of stainless steel ($\frac{1}{2}$ in. thick).

In order to convert observed counting rates to absolute activities, the counting efficiency of the beta-proportional counter was determined by means of self-absorption curves obtained for 4-π counted samples of five of the longer-lived nuclides using the method suggested by Steinberg.[20] Such a technique is exact only where the radioactive species to be counted has a relatively long half-life so that a self-absorption curve may be measured. For short-lived nuclides, an alternative method outlined by Bayhurst and Prestwood[21] was used for determining detection efficiencies.

The gamma-ray spectra were used as a check on the relative amounts of the activities of various nulcides by comparing areas under the photopeaks divided by the peak-to-total ratios as given by Heath.[22]

For both beta and gamma counting, branching ratios were taken from Nuclear Data Sheets[23] and from Katcoff.[24]

Treatment of Data

For each sample, the gross beta decay was observed and resolved into the various components using reported half-lives.

From a knowledge of the absolute activity at the end of the irradiation after correcting for parent-daughter growths and decays, branching decays, and counting efficiency, the total number of atoms of a particular species produced during the irradiation was determined by means of the following equation which corrects for the decay of the species during the irradiation:

$$A_{\text{total}}=[A\lambda A/(1-e^{-\lambda A t})]t, \qquad (1)$$

where A_{total} is the total number of atoms of species A produced during the irradiation, $A\lambda A$ is the activity of species A at the end of the irradiation, and t is the length of the bombardment.

The absolute yield of Mo^{99} was determined for the 14.8-MeV irradiations by using an aluminum monitor and for the 2.95-MeV irradiations by using a gold monitor. Then, all of the other yields were measured relative to Mo^{99} by means of the following equation:

$$\text{Yield of } A=\frac{\text{Yield of } Mo^{99}}{Mo^{99}\text{ total}}(A_{\text{total}}). \qquad (2)$$

Sources of Error

(1) An error in fitting the decay curve to the measured activity introduces an uncertainty in the total number of atoms produced by the irradiation. Such error amounted to $\sim 3\%$ for the longer-lived nuclides and $\sim 5\%$ for the short-lived species.

(2) Corrections were made for fluctuations in the neutron flux during the irradiations where possible, and in other cases agreement between triplicate samples was required.

(3) Errors introduced by thermal neutrons, 2.95-MeV neutrons produced during 14.8-MeV neutron irradiations, or chemical yield determinations were insignificant.

The errors reported for all the yields measured in this work are most probable errors propagated in the usual way, assuming the Mo^{99} yield to be 1.96% at 14.8 MeV and 3.10% at 2.95 MeV as measured in this work.

Absolute Fission Yield of Mo^{99}

Mo^{99} was chosen as the reference nuclide for this investigation. In order to convert the relative yields of the other nuclides to absolute yields, the Mo^{99} was measured using the $Al^{27}(n,\alpha)Na^{24}$ cross section of 115 mb as reported by Poularikas and Fink[25] for the 14.8-MeV irradiations. Similarly, the $Au^{197}(n,\gamma)Au^{198}$ cross of 49 mb as reported by Greisen[26] was used for the 2.95-MeV irradiations.

A sample of $Th(NO_3)_4\cdot 2H_2O$ was "sandwiched" between double layers of aluminum or gold foil and irradiated with 14.8- or 2.95-MeV neutrons, respectively, for two hours. In each case, Mo^{99} was isolated after allowing the short-lived molybdenum isotopes to decay.

By counting both the Mo^{99} and the monitor foil, the activity of the active species at the end of the irradiation was obtained. Since these nuclides have different half-lives, it was necessary to correct these data to saturation bombardment time.

Using the thin target approximation,[27] the cross section for the reaction may be obtained from

$$\sigma_{\text{sample}}=\frac{N_{\text{sample}}Q_{\text{monitor}}}{N_{\text{monitor}}Q_{\text{sample}}}\cdot\sigma_{\text{monitor}} \qquad (3)$$

since both monitor and sample were of equal area and were irradiated under identical conditions. Here, σ is the cross section for the process in question (cm^2); N is the absolute activity for saturation bombardment; and Q is the number of target nuclei per cm^2. Since all of the terms on the right-hand side of the equation are known, σ_{sample} may be calculated.

This technique was employed to calculate the abso-

[20] E. P. Steinberg, ANL-5622, 1956 (unpublished).
[21] B. P. Bayhurst and R. J. Prestwood, Nucleonics **17**, No. 3, 82 (1959).
[22] R. L. Heath, IDO-16408, 1957 (unpublished).
[23] *Nuclear Data Sheets* (National Academy of Sciences–National Research Council, Washington, D. C., 1962).
[24] S. Katcoff, Nucleonics **18**, No. 11, 201 (1960).
[25] A. Poularikas and R. W. Fink, Phys. Rev. **115**, 989 (1959).
[26] K. I. Greisen, MDDC-3, 1944 (unpublished).
[27] G. Friedlander and J. Kennedy, *Nuclear and Radiochemistry*, (John Wiley & Sons, Inc., New York, 1955), pp. 60–62.

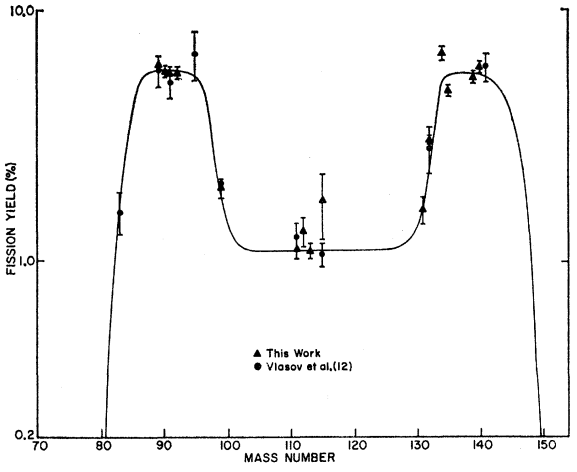

FIG. 1. Th^{232} 14-MeV neutron fission mass yield curve.

lute yields of Mo^{99} by assuming values for the neutron-induced fission cross section of Th^{232} to be 0.37 b at[28] 14.8 MeV and 0.17 b at[29] 2.95 MeV.

The results of two such determinations for each neutron energy employed gave values of $5.27(\pm 0.12)\times 10^{-3}$ b and $7.25(\pm 0.55)\times 10^{-3}$ b for the partial fission cross section of Th^{232} leading to mass number 99 for 2.95- and 14.8-MeV neutrons, respectively. Converting to fission yields (%), these values give $3.10\pm 0.11\%$ and $1.96\pm 0.15\%$ for the fission yields of Mo^{99} at 2.95 and 14.8 MeV, respectively.

III. RESULTS

Yields of Strontium Isotopes

Yields of strontium isotopes 89–92 were obtained from three irradiations at 14.8 MeV and two irradiations at 2.95 MeV. The 14.8-MeV yield for Sr^{90} was obtained by allowing samples to decay for 1.4 years, redissolving the samples and separating Sr^{90} from Y^{90} by carrying the Y^{90} on $Fe(OH)_3$.

In the case of the 2.95-MeV irradiations, the activities of 50.4-day Sr^{89} and 28-year Sr^{90} were too low to be observed. Hence, only the shorter-lived nuclides could be measured.

Yields of Silver Isotopes

Using radiochemical separation techniques, silver samples were isolated and the yields of Ag^{111}, Ag^{112}, Ag^{113}, and Ag^{115} were calculated from two irradiations with 14.8-MeV neutrons. Results of two irradiations with 2.95-MeV neutrons gave the yield for Ag^{113}.

Yields of Iodine Isotopes

Samples of iodine were separated by standard methods at varying lengths of time after the irradiations. For all of the samples both gross-beta decay and gamma-ray spectra were obtained. Since most of the iodine nuclides have radioactive-xenon daughters, sealed samples were used in order to avoid corrections for outgassing of the xenon. The yields were calculated from three sets of samples at each of the two neutron energies.

Yields of Barium Isotopes

Similarly, yields were caluclated for Ba^{139} and Ba^{140} from two irradiations at 14.8 MeV and for Ba^{139} from two irradiations at 2.95 MeV using samples separated by standard methods. While all the above yields are reported as "yield of Sr^{90}," etc., the values given have been corrected where necessary for holdup in the decay chain by a long-lived precursor and actually represent cumulative fission yields.

The values of the yields measured in this work as well as those determined by Vlasov *et al.*[12] for 14.3-MeV neutrons, Turkevich, Nidday, and Tompkins[30] for 6–11-MeV neutrons, and Turkevich and Nidday[13] for 2.6-MeV neutrons are listed in Table I and are plotted in Figs. 1, 2, and 3.

IV. DISCUSSION

General Features of the Mass Yield Curves

In general certain effects are expected in all mass yield curves. The peak-to-valley ratio of the mass yield curve is expected to decrease with increasing energy of the incident particle. In the present investigation this was observed. The peak-to-valley ratio of 5.0 for 14-MeV neutron fission is a factor of 28.6 lower than the peak-to-valley ratio of 143 for 2.95-MeV neutron fission. This is a reasonable decrease when compared to U^{238} neutron-induced fission,[9] where the peak-to-valley ratios are 7.15 and 200 for 14- and 2.6-MeV neutron fission. The decrease in this case is a factor of 28.0 which agrees well with 28.6 for the present data.

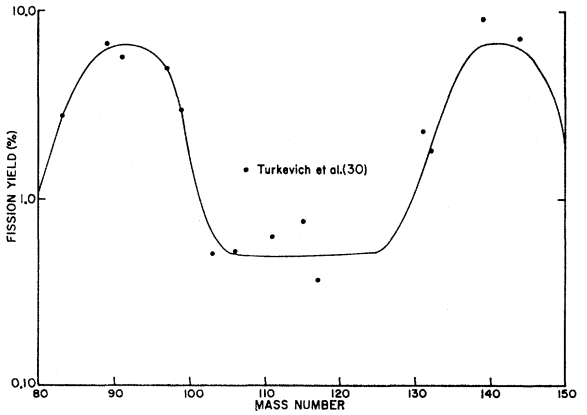

FIG. 2. Th^{232} 6–11-MeV neutron fission mass yield curve.

[28] J. H. Williams, LA-520, 1946 (unpublished).
[29] A. A. Berezin, G. A. Stol'arov, Y. V. Nikol'skii, and I. E. Chelnokov, At. Energ. 5, 659 (1958).
[30] A. Turkevich, J. B. Nidday, and A. Tompkins, Phys. Rev. 89, 552 (1953).

TABLE I. Th^{232} fast neutron fission yields.

Nuclide	14.8 MeV This work (%)	14.3 MeV Vlasov *et al.* Ref. 12. (%)	2.95 MeV This work (%)	2.6 MeV Turkevich *et al.* Ref. 13. (%)	6 to 11 MeV Turkevich *et al.* Ref. 30 (%)
Zn^{72}				0.00033±0.00008	
Ga^{73}		<0.06		0.00045±0.00022	
Ge^{77}				0.020 ±0.007	0.052
Br^{83}		1.6 ±0.3		1.9 ±0.45	2.74
Sr^{89}	6.03±0.48	5.7 ±0.8		6.7 ±0.7	6.7
Sr^{90}	5.72±0.81			6.1 ±1.2	
Sr^{91}	5.52±0.52		6.40 ±0.20	6.4 ±0.7	5.6
Y^{91}		5.2 ±0.8			
Sr^{92}	5.58±0.53		6.60 ±0.28		
Zr^{95}		6.7 ±1.5			
Zr^{97}				5.4 ±0.8	4.95
Mo^{99}	1.96±0.15	2.0 ±0.2	3.10 ±0.11	2.9 ±0.3	3.1
Ru^{103}				0.20 ±0.07	0.51±0.25
Rh^{105}				0.07 ±0.02	
Rh^{106}				0.058 ±0.006	0.53
Pd^{109}				0.053 ±0.010	
Ag^{111}	1.13±0.11	1.27±0.15		0.052 ±0.010	0.63
Ag^{112}	1.32±0.17				
Pd^{112}				0.065 ±0.010	
Ag^{113}	1.10±0.08		0.047±0.009		
Ag^{115}	1.72±0.50				
Cd^{115}		1.07±0.12		0.075 ±0.015	0.76
Cd^{117}					0.37±0.18
I^{131}	1.59±0.21		1.15 ±0.14	1.2 ±0.6	2.3
I^{132}	3.10±0.15		2.50 ±0.19		
Te^{132}		2.8 ±0.6		2.4 ±0.7	1.8
I^{133}	3.78±0.18		3.26 ±0.31		
I^{134}	6.69±0.36		8.15 ±0.92		
I^{135}	4.74±0.24		5.57 ±0.60		
Cs^{139}				6.6 ±1.0	
Ba^{139}	5.34±0.37		6.78 ±0.50		9.0
Ba^{140}	5.97±0.35			6.2 ±2.0	
Ce^{141}		5.9 ±0.8		9.0 ±3.0	
Ce^{144}				7.1 ±1.0	7.2

This effect is interpreted as evidence for two separate, distinct fission modes, symmetric and asymmetric.[13] As the energy of the incident particle is increased, the ratio of the probability of symmetric fission to that of asymmetric fission increases, and hence the peak-to-valley ratio decreases. At an incident particle energy of approximately 50 MeV, the double-humped curve disappears completely, and one broad curve corresponding to symmetric fission is observed.[31]

Very qualitatively this effect may be explained by postulating that an inner core of roughly forty nucleons (20 proton and neutron closed shells with perhaps some contribution by the 28 and 50 proton and neutron closed shells) remains relatively undisturbed during the liquid-drop oscillations leading to fission at low-excitation energies. Thus, asymmetric mass division predominates at low energies with a difference in fragment masses of approximately forty. At higher energies this inner core becomes ruptured more and more frequently and at sufficiently high excitation energies loses its influence entirely. Thus, symmetric fission becomes most probable.

The position of the maximum in the heavy mass peak is expected to remain relatively unchanged with respect to mass number while the peak height is expected to decrease slightly with increasing incident particle energy. Such is the case for Th^{232} since the maximum in the heavy mass peak remains at about 138 for both 2.96- and 14.8-MeV neutron fission while the maximum value of the peak decreases from 7.2 to 5.6 percent as the energy increases from 2.95- to 14.8-MeV.

The maximum in the light mass peak is generally observed to shift slightly towards higher masses as the

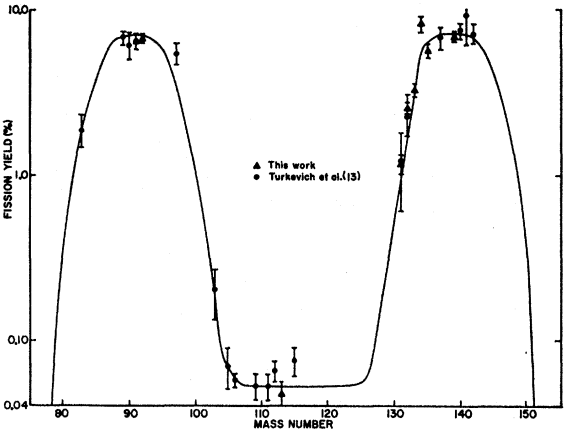

FIG. 3. Th^{232} 3-MeV neutron fission mass yield curve.

[31] R. A. Schmitt and N. Sugarman, Phys. Rev. **95**, 1260 (1954).

mass number of the target nucleus increases. For a given target nucleus, however, its maximum is expected to remain constant with respect to mass number while its maximum value is expected to decrease as the energy of the incident particle increases. These effects are also observed since the maximum value decreases from about 7.0% to 5.6% as the neutron energies increase from 2.95 to 14.8 MeV, respectively. The position of the peak remains relatively constant with a maximum value for $A=92$ for both neutron energies while comparison with U^{235} fission yields indicates that as mass number of the target nucleus decreases from 235 to 232 the maximum shifts from 95 to 92. The shifting of the light mass peak to higher mass numbers as the mass of the target nucleus is increased can be explained qualitatively from the fact that the mass of the light fragment A_L is roughly equal to the difference between the mass of the fissioning nuclide A_T and the mass of the corresponding heavy fragment A_H. Therefore, for a given A_H, as A_T increases, A_L increases correspondingly. A somewhat more sophisticated explanation of this effect was offered by Swiatecki.[32]

The wings of the peaks should be observed to splay out slightly with increasing excitation energy. As seen in Fig. 1 and Fig. 3, the peak width at half-maximum increases from 13 for 2.95 MeV to 14 for 14.8 MeV.

The agreement between the values determined in this work and the values determined by other investigators for neutron-induced fission at[12] 14.2 MeV and at[13] 2.6 MeV is considered to be good. At every point (except that for $A=115$ for 14-MeV fission) for which the present data and the other determinations coincide, the values agree within the reported error limits. For the case of 14-MeV neutron fission, the yield for $A=115$ reported in this work is slightly higher than that reported by Vlasov *et al.*,[12] but since the errors are admittedly large and the disagreement is not large, perhaps this discrepancy is understandable.

Fine Structure in the Mass Yield Curves

One point of interest exhibited by the present data for 14-MeV neutron-induced fission of Th^{232} is the fine structure in the region $A=131$–135. It should be noted that the yield at $A=132$ is slightly higher than the smooth curve, while the value of $A=134$ is approximately thirty percent higher than would be expected from the smooth curve drawn through the neighboring yields. This effect was also noted in data from 2.95-MeV neutron fission. The same pattern is observed with the peak at $A=134$ being about fifty percent higher than expected. Although in this case the error limits are large, the minimum error limit is more than thirty-five percent in excess of the value predicted by the smooth curve.

In order to explain this fine structure, attempts were made to calculate predicted fission yields in the mass range $A=131$–135. The first method tried was that of Glendenin.[33] First, from the postulate of equal charge displacement by Glendenin, Coryell, and Edwards,[34] Z_P, the most probable charge for any given mass chain, was calculated by means of the following equation:

$$Z_P=Z_A-\tfrac{1}{2}(Z_A+Z_{233-A-\nu}-Z_{233}), \qquad (4)$$

where Z_A is the most stable charge for any mass chain A, $Z_{233-A-\nu}$ is the most stable charge for the mass chain complementary to the fragment of mass A, and Z_{233} is the number of protons in the target nucleus, i.e., $Z_{Th^{232}}=90$. The values of Z_A and Z_{233-A} were taken from Coryell.[35] These values differ from some used in early work on fission in that they include corrections for the discontinuities in the smooth curve of Z_A versus A which appear at closed shell edges.

The results of these caluclations are shown in Table II.

TABLE II. Values of Z_p calculated by Glendenin's method, Ref. 34.

A	233-A	Z_A	Z_{233-A}	Z_p
131	102	54.6	44.6	50.00
132	101	54.8	44.2	50.30
133	100	55.4	44.0	50.70
134	99	55.6	43.7	50.95
135	98	55.8	43.3	51.25
136	97	56.3	42.8	51.75

Next, the fission yields which would be expected if no fine structure were present for the mass numbers in question, were determined by normalizing the fission yield curves to 200%. (See smooth curve in Figs. 1 and 3.) The values which were used are given in Table III.

TABLE III. Fission yields taken from smooth curves in Figs. 1 and 3.

A	Yield from 2.95-MeV fission (%)	Yield from 14.8-MeV fission (%)
131	0.9	1.5
132	1.7	2.4
133	3.1	3.6
134	5.5	5.2
135	6.4	5.4
136	6.7	5.5

In order to determine the primary yield distribution along each mass chain, the charge distribution curve determined by Ford *et al.*[36] was used.

[32] W. J. Swiatecki, Phys. Rev. **100**, 936 (1955).

[33] L. E. Glendenin, Phys. Rev. **75**, 337 (1949).

[34] L. E. Glendenin, C. D. Coryell, and R. R. Edwards, in *Radiochemical Studies: The Fission Products*, (McGraw-Hill Book Company, Inc., New York, 1951), Book 1, Paper 52.

[35] C. D. Coryell, Ann. Rev. Nucl. Sci. **2**, 325 (1953).

[36] G. P. Ford, J. S. Gilmore, D. P. Ames, J. P. Balagna, J. W. Barnes, A. A. Comstock, G. A. Cowan, P. B. Elkin, D. C. Hoffman, G. W. Knobeloch, E. J. Lang, M. A. Melnick, C. O. Minkkinen, B. D. Pollock, J. E. Sattizshn, C. W. Stanley, and B. Warren, LA-1997, 1956 (unpublished).

From the values of Z_p and the cumulative yields, the primary yields were calculated and are given in Table IV and Fig. 4.

TABLE IV. Primary fission yields from Glendenin's method, Ref. 33.

					Cumulative yield %		Primary yield %	
A	Z	Z_p	$Z-Z_p$	Fractional yield	2.95 MeV	14.8 MeV	2.95 MeV	14.8 MeV
131	52	50.00	2	0.022	0.90	1.52	0.020	0.033
	51	50.00	1	0.26	0.90	1.52	0.234	0.395
	50	50.00	0	0.49	0.90	1.52	0.441	0.745
	49	50.00	−1	0.26	0.90	1.52	0.234	0.395
	48	50.00	−2	0.022	0.90	1.52	0.020	0.033
132	52	50.30	1.7	0.07	1.7	2.4	0.119	0.168
	51	50.30	0.7	0.36	1.7	2.4	0.612	0.863
	50	50.30	−0.3	0.44	1.7	2.4	0.748	1.057
	49	50.30	−1.3	0.17	1.7	2.4	0.289	0.408
	48	50.30	−2.3	0.0017	1.7	2.4	0.003	0.004
133	53	50.70	2.3	0.0017	3.1	3.6	0.005	0.006
	52	50.70	1.3	0.17	3.1	3.6	0.527	0.612
	51	50.70	0.3	0.44	3.1	3.6	1.363	1.583
	50	50.70	−0.7	0.36	3.1	3.6	1.117	1.297
	49	50.70	−1.7	0.07	3.1	3.6	0.217	0.252
134	53	50.95	2.05	0.017	5.5	5.2	0.094	0.088
	52	50.95	1.05	0.25	5.5	5.2	1.374	1.299
	51	50.95	0.05	0.49	5.5	5.2	2.696	2.545
	50	50.95	−0.95	0.25	5.5	5.2	1.374	1.299
	49	50.95	−1.95	0.016	5.5	5.2	0.088	0.083
135	53	51.25	1.75	0.052	6.4	5.4	0.333	0.281
	52	51.25	0.75	0.33	6.4	5.4	2.113	1.782
	51	51.25	−0.25	0.47	6.4	5.4	3.003	2.537
	50	51.25	−1.25	0.16	6.4	5.4	1.023	0.863
	49	51.25	−2.25	0.0035	6.4	5.4	0.022	0.019
136	54	51.75	2.25	0.0035	6.7	5.5	0.023	0.019
	53	51.75	1.25	0.16	6.7	5.5	1.072	0.879
	52	51.75	0.25	0.47	6.7	5.5	3.150	2.582
	51	51.75	−0.75	0.33	6.7	5.5	2.212	1.815
	50	51.75	−1.75	0.052	6.7	5.5	0.348	0.286

Glendenin[33] proposed that the fine structure arises from the fact that since the eighty-third neutron is weakly bound, it may be emitted in addition to the usual number of prompt neutrons emitted by each fragment. To compensate for this effect, the primary yields for the nuclides having eighty-three neutrons were transferred to those having eighty-two neutrons as indicated by the arrows in Fig. 4. Next, the cumulative yields were obtained by summing the primary yields for each mass chain. The results are presented in Table V, with the experimental yields shown for comparison.

The disagreement between the calculated values and the experimental values is obvious with the large difference at $A=134$ being even lower than the values taken from the smooth curve.

The next method tried for the explanation of the experimental data was proposed by Pappas.[37] He suggested two refinements of the method proposed by Glendenin. First, he stated that the postulate of equal charge displacement should apply to the fragments before the emission of prompt neutrons. Second, due to binding energy systematics, he suggested that the extra boil-off of neutrons should be extended to include the third, fifth, and perhaps even the seventh neutron beyond a closed neutron shell. Calculation of Z_p by Pappas' method is accomplished by means of the modified form of Eq. 4

$$Z_p=Z_{A+n}-\tfrac{1}{2}(Z_{233-A-n}+Z_{A+n}-Z_{233}), \qquad (5)$$

where Z_{A+n} is the most stable charge for any mass chain $A+n$, $Z_{233-A-n}$ is the most stable charge for the complementary fragment mass chain, Z_{233} is 90, A is the mass number in question, and n is the average number ($\bar{\nu}_H$) of neutrons emitted by the heavy fragment.

In using this method for the present investigation, however, one difficulty arises. Pappas' method requires a knowledge of the average number of prompt neutrons emitted by each fragment. In some cases this has been determined,[9] but such data are not available for Th^{232}. However, $\bar{\nu}$, the average total number of neutrons emitted per Th^{232} fission has been measured,[38] and values of 2.42 ± 0.10 and 4.43 ± 0.13 for 3.6- and 14.9-MeV neutrons, respectively, were reported. Since these data correspond closely to those for U^{238}, we may assume that $\bar{\nu}_H$, the average number of neutrons emitted by the heavy fragment, is equal to three for 14.8-MeV fission since that is the value observed for U^{238}. Likewise, we can assume a value of $\bar{\nu}_H$ of one corresponding to that for U^{238} for 2.95-MeV fission. Although these assumptions are somewhat arbitrary, still a qualitative explanation of the observed yields can be expected since the method of Pappas is only expected to yield qualitative results.

The values of Z_p calculated from Eq. (5) for $\bar{\nu}_H=3$ for 14.8-MeV fission and $\bar{\nu}_H=1$ for 2.95-MeV fission are given in Table VI. The value for the fission yields expected in the absence of fine structure were taken from the smooth curve in Fig. 1 and Fig. 3 as before (see Table III), and using the charge distribution curve of Ford[36] the primary fission yields were calculated and are given in Table VII and Fig. 5.

TABLE V. Cumulative yields calculated by Glendenin's method, Ref. 33 and experimental cumulative yields.

	2.95-MeV cumulative yields		14.8-MeV cumulative yields	
A	Calculated (%)	Experimental (%)	Calculated (%)	Experimental (%)
131	1.17	1.15	1.90	1.59
132	2.53	2.50	3.29	3.10
133	4.68	3.26	4.85	3.78
134	4.92	8.15	4.44	6.69
135	5.36	5.57	4.50	4.74

[37] A. C. Pappas, MIT-Tech Report No. 63, September 1953 (unpublished).

[38] H. Conde and N. Starfelt, Nucl. Sci. Engr. **11**, 397 (1961).

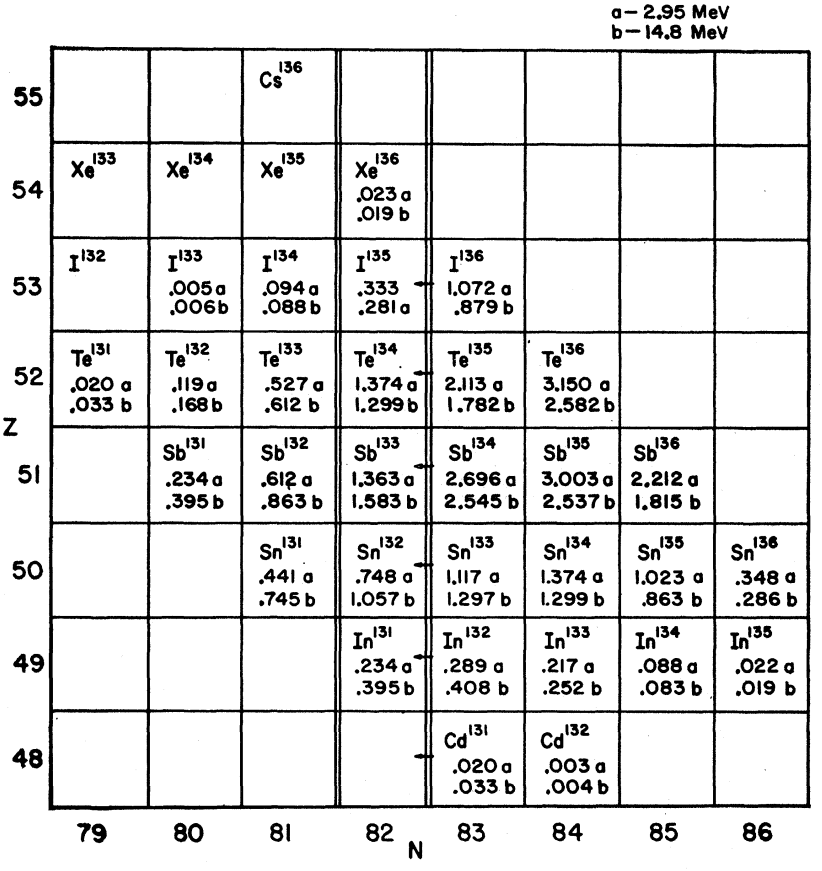

FIG. 4. Primary yields calculated by Glendenin's method (Ref. 33).

According to Pappas' postulate the fine structure is due to excess neutron emission by nuclides having 83, 85, and 87 neutrons. For the present data only the 83 and 85 neutron cases need be considered. (As will be seen later, the primary yields for nuclides having 87 neutrons are nearly zero for the cases considered here.) By transferring these primary yields as indicated by the arrows in Fig. 5 and summing the primary yields, the calculated cumulative yields were obtained. These are shown in Table VIII with the experimental data for comparison. This treatment predicts the shape of the fine structure approximately.

TABLE VI. Values of Z_p calculated by Pappas' method (Ref. 37).

n	A	A_{Z+n}	$A_{233-A-n}$	Z_{A+n}	$Z_{233-A-n}$	Z_p
3	131	134	99	55.6	43.7	50.95
	132	135	98	55.8	43.3	51.25
	133	136	97	56.3	42.8	51.75
	134	137	96	56.7	42.2	52.25
	135	138	95	57.1	41.9	52.60
	136	139	94	57.6	41.3	53.15
1	131	132	101	54.8	44.2	50.30
	132	133	100	55.4	44.0	50.70
	133	134	99	55.6	43.7	50.95
	134	135	98	55.8	43.3	51.25
	135	136	97	56.3	42.8	51.75
	136	137	96	56.7	42.2	52.25

An additional refinement to Pappas' method has been proposed. As pointed out by the author,[9] an additional effect should be considered in the extra neutron boil-off by nuclides beyond a closed shell. As the mass number is increased at a constant neutron number in the region under consideration above, the nuclide configuration approaches stability. For the isotones of $N=83$ when A is increased from 131 up to 134 or 136, i.e., Sb^{134} and I^{136}, sufficient stability has been achieved for the nuclides to have measurable beta decay half-lives of 45 and 86 sec, respectively. These nuclides should be expected to emit extra neutrons less frequently than other 83-neutron isotones further from stability. We may exaggerate this effect by assuming that these particular species do not emit neutrons at all and see what effect this has upon the data. The results are shown in Fig. 6 with the data from Table VIII.

The corrected data predict more accurately the shape of the observed fine structure, but it is obvious that the reduction in extra neutron emission by Sb^{134} and I^{136} of 100% is too large. If we now choose a reduction factor of 50 percent (i.e., assume that due to the added stability of Sb^{134} and I^{136} these species emit their eighty-third neutron only half as frequently as other less stable isotones), we obtain the data given in Fig. 7. The agreement is considered surprisingly good when it is noted that the calculations are not expected to be quantitative.

TABLE VII. Primary yields calculated by Pappas' method (Ref. 37).

A	Z	Z_p	$Z-Z_p$	Fractional yield	Cumulative yield (%)	Primary yield (%)
		$n=3$ (i.e., 14.8-MeV neutron fission)				
131	53	50.95	2.05	0.017	1.52	0.026
	52	50.95	1.05	0.25	1.52	0.380
	51	50.95	0.05	0.49	1.52	0.745
	50	50.95	−0.95	0.25	1.52	0.380
	49	50.95	−1.95	0.016	1.52	0.024
132	53	51.25	1.75	0.052	2.4	0.125
	52	51.25	0.75	0.33	2.4	0.792
	51	51.25	−0.25	0.47	2.4	1.128
	50	51.25	−1.25	0.16	2.4	0.384
	49	51.25	−2.25	0.0035	2.4	0.008
133	54	51.75	2.25	0.0035	3.6	0.013
	53	51.75	1.25	0.16	3.6	0.576
	52	51.75	0.25	0.47	3.6	1.691
	51	51.75	−0.75	0.33	3.6	1.188
	50	51.75	−1.75	0.052	3.6	0.187
134	54	52.25	1.75	0.052	5.2	0.270
	53	52.25	0.75	0.33	5.2	1.718
	52	52.25	−0.25	0.47	5.2	2.445
	51	52.25	−1.25	0.16	5.2	0.832
	50	52.25	−2.25	0.0035	5.2	0.018
135	55	52.60	2.40	0.0015	5.4	0.008
	54	52.60	1.40	0.14	5.4	0.756
	53	52.60	0.40	0.42	5.4	2.267
	52	52.60	−0.60	0.39	5.4	2.105
	51	52.60	−1.60	0.095	5.4	0.513
136	55	53.15	1.85	0.028	5.5	0.154
	54	53.15	0.85	0.26	5.5	1.430
	53	53.15	−0.15	0.46	5.5	2.530
	52	53.15	−1.15	0.24	5.5	1.320
	51	53.15	−2.15	0.015	5.5	0.082
		$n=1$ (i.e., 2.95-MeV neutron fission)				
131	52	50.30	1.70	0.07	0.90	0.063
	51	50.30	0.70	0.36	0.90	0.324
	50	50.30	−0.30	0.44	0.90	0.396
	49	50.30	−1.30	0.17	0·90	0.153
	48	50.30	−2.30	0.0017	0.90	0.001
132	53	50.70	2.30	0.0017	1.7	0.003
	52	50.70	1.30	0.17	1.7	0.289
	51	50.70	0.30	0.44	1.7	0.748
	50	50.70	−0.70	0.36	1.7	0.612
	49	50.70	−1.70	0.07	1.7	0.119
133	53	50.95	2.05	0.017	3.1	0.053
	52	50.95	1.05	0.25	3.1	0.775
	51	50.95	0.05	0.49	3.1	1.519
	50	50.95	−0.95	0.25	3.1	0.775
	49	50.95	−1.95	0.016	3.1	0.050
134	53	51.25	1.75	0.052	5.5	0.286
	52	51.25	0.75	0.33	5.5	1.815
	51	51.25	−0.25	0.47	5.5	2.585
	50	51.25	−1.25	0.16	5.5	0.879
	49	51.25	−2.25	0.0035	5.5	0.019
135	54	51.75	2.25	0.0035	6.4	0.022
	53	51.75	1.25	0.16	6.4	1.024
	52	51.75	0.25	0.47	6.4	3.010
	51	51.75	−0.75	0.33	6.4	2.113
	50	51.75	−1.75	0.052	6.4	0.333
136	54	52.25	1.75	0.052	6.7	0.348
	53	52.25	0.75	0.33	6.7	2.212
	52	52.25	−0.25	0.47	6.7	3.150
	51	52.25	−1.25	0.16	6.7	1.072
	50	52.25	−2.25	0.0035	6.7	0.023

TABLE VIII. Cumulative yields calculated by Pappas' method (Ref. 37) and experimental cumulative yields.

	2.95-MeV cumulative yields		14.8-MeV cumulative yields	
A	Calculated (%)	Experimental (%)	Calculated (%)	Experimental (%)
131	1.02	1.15	1.53	1.59
132	2.36	2.50	2.58	3.10
133	4.93	3.26	4.24	3.78
134	6.24	8.15	6.47	6.69
135	6.34	5.57	5.91	4.47

Another method for calculating fission yield fine structure has been suggested recently by Terrell.[39] This method makes use of both primary fission yields, cumalative fission yields and the number of neutrons emitted as a function of mass number. Unfortunately, only the cumulative yields are known for Th^{232} at the present time so that any application of Terrell's method is impossible for the present investigation.

Fission Yield Fine Structure and the Terrestrial Abundance of the Xenon Isotopes

Fine structure in the mass yield curves has been reported for the fission of Th^{232}, U^{233}, U^{235}, U^{238}, Pu^{239}, and Cm^{242}.[40] One point of interest is that the peak in the fine structure occurs for $A=134$ in every case except those of U^{233} and U^{238}.[40,9] For U^{233} and U^{238} the peak in the fine structure occurs for $A=133$ and $A=132$, respectively.

When the neutron excess, $N-Z$, is plotted as a function of mass number, a series of straight lines is obtained as seen in Fig. 8. It is noteworthy that all of the nuclides mentioned above as having peaks in their fine structure at $A=134$ have a relatively constant value of $N-Z$. U^{238} lies considerably above this line and has a value of $N-Z$ corresponding closely to that for Pu^{244} and Cm^{247}. It is interesting to speculate that the nuclides having the same value of $N-Z$ as does U^{238} (i.e., Pu^{244} and Cm^{247}) might exhibit the same fine structure as that observed for U^{238}; namely, a peak at $A=132$. Of course, it would be desirable to calculate the fine structure based on the methods used in the preceding section. However, any such calculations require a knowledge of at least some points on the fission yield curve and a knowledge of the number of prompt neutrons emitted per fission event, ν. Unfortunately, neither of these has been determined for Pu^{244} or for Cm^{247}. An extremely rough estimate of the fine structure, based on the assumptions that ν is similar in each case to that for Cf^{252} spontaneous fission[41] and that the smooth curve remains constant at six percent for $A=131$–135, does indicate a peak in the fine structure at $A=132$.

[39] J. Terrell, Phys. Rev. **127**, 880 (1962).
[40] E. K. Hyde, UCRL-9036-Rev., 1962 (unpublished).
[41] L. E. Glendenin and E. P. Steinberg, J. Inorg. Nucl. Chem. **1**, 45 (1955).

a–2.95 MeV
b–14.8 MeV

Z \ N	79	80	81	82	83	84	85	86
55		Cs^{136} .008 a	Cs^{136} .154 a .010 b					
54	Xe^{133} .013 a	Xe^{134} .270 a	Xe^{135} .756 a .022 b	Xe^{136} 1.430 a .938 b				
53	I^{132} .125 a .003 b	I^{133} .576 a .053 b	I^{134} 1.718 a .268 b	I^{135} 2.267 a 1.024 b	I^{136} (86 sec) 2.530 a 2.212 b			
52	Te^{131} .380 a .063 b	Te^{132} .792 a .289 b	Te^{133} 1.691 a .775 b	Te^{134} 2.445 a 1.815 b	Te^{135} 2.105 a 3.010 b	Te^{136} 1.320 a 3.150 b		
51		Sb^{131} .745 a .396 b	Sb^{132} 1.128 a .768 b	Sb^{133} 1.188 a 1.519 b	Sb^{134} (45 sec) .832 a 2.585 b	Sb^{135} .513 a 2.113 b	Sb^{136} .082 a 1.072 b	
50			Sn^{131} .380 a .324 b	Sn^{132} .384 a .612 b	Sn^{133} .187 a .775 b	Sn^{134} .018 a .879 b	Sn^{135} .022 b	Sn^{136} .023 b
49				In^{131} .024 a .153 b	In^{132} .008 a .119 b	In^{133} .050 b	In^{134} .019 b	
48					Cd^{131} .001 b			

FIG. 5. Primary yields calculated by Pappas' method (Ref. 37).

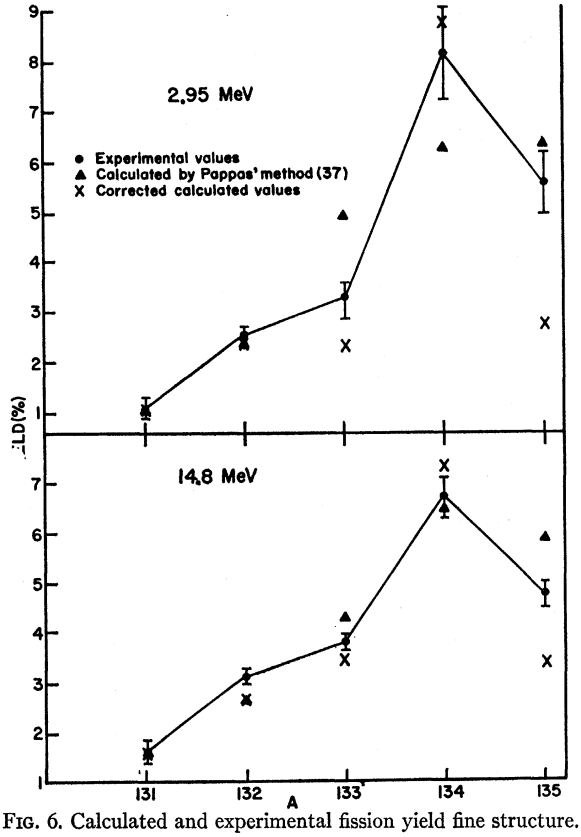

FIG. 6. Calculated and experimental fission yield fine structure.

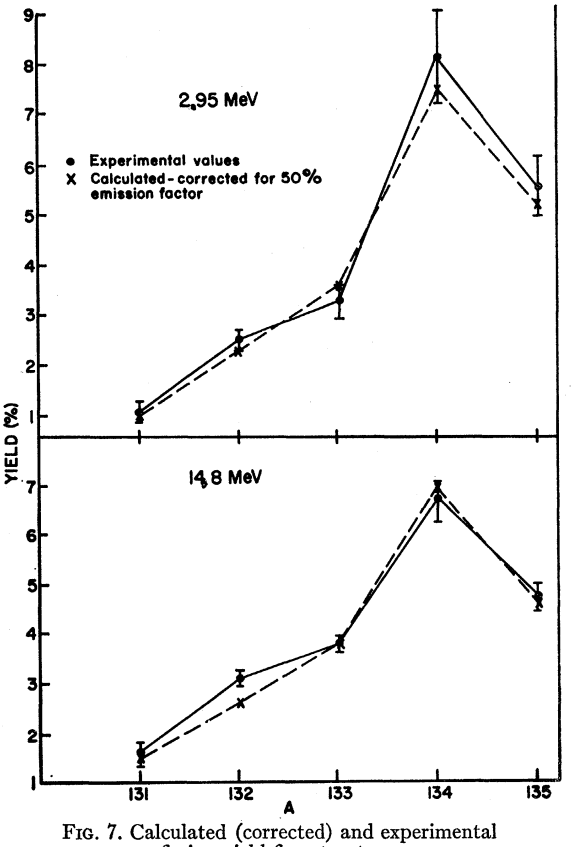

FIG. 7. Calculated (corrected) and experimental fission yield fine structure.

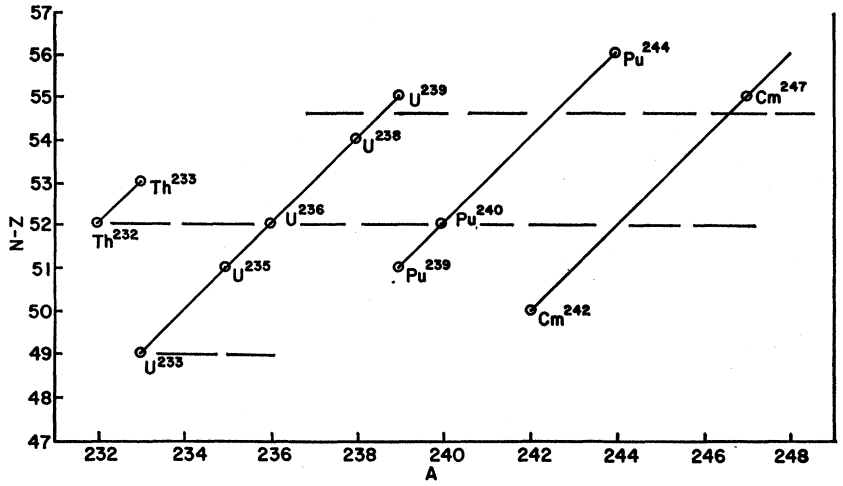

FIG. 8. Neutron excess versus mass number for heavy elements.

If such a trend were in fact correct, the suggestion made by Kuroda[42] should be considered. Reynolds has shown that there is an excess of Xe^{132} in the isotopic abundances of terrestrial xenon isotopes.[43] Kuroda proposed that this excess is due to the spontaneous fission of some extinct transuranium isotope. His candidate for the source of the excess Xe^{132} is Pu^{244} since it has a half-life of 7×10^7 years and thus could have survived the interval of approximately 10^8 years from the cessation of element synthesis to the formation of the earth. If the above-mentioned trend in fission yield fine structure is correct, this proposal seems quite logical. Also, recent work has shown Cm^{247} to have a half-life of approximately 10^8 years,[44,45] and it follows that this nuclide might also have contributed to the excess Xe^{132}.

As mentioned above, the peak in the fine structure observed for U^{233} photofission appears at $A=133$. All attempts at explanations of this effect by the methods which predict fine structure in other cases have failed. However, it should be noted that there is some disagreement in the literature concerning the existence of the peak at $A=133$. Although Steinberg *et al.*[46] reported the peak, Thode[47] could find no evidence for fine structure for $A=131$–136.

At the present time in this laboratory, investigations are being made of photofission reactions in an attempt to resolve the apparently anomalous results.

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[42] P. K. Kuroda, Nature **187**, 36 (1960).
[43] J. H. Reynolds, Phys. Rev. Letters **4**, 8 (1960).
[44] V. V. Cherdyntsev and V. F. Mikhailov, Geokhim. **1**, 3 (1963).
[45] M. Nurmia (private communication, 1963).
[46] E. P. Steinberg, L. E. Glendenin, M. G. Inghram, and R. J. Hayden, Phys. Rev. **95**, 867 (1954).
[47] R. K. Wanless and H. G. Thode, Can. J. Phys. **33**, 541 (1955).